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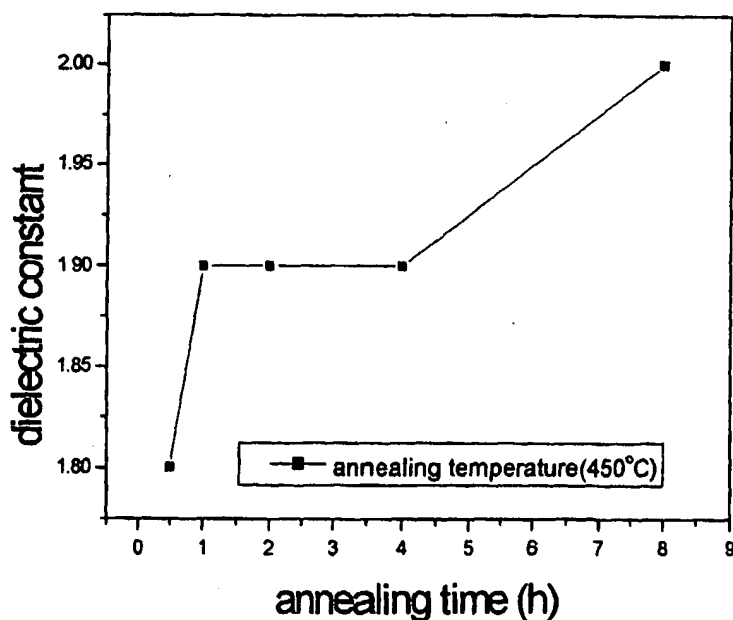
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(54) Title: **METHOD FOR PREPARING LOW DIELECTRIC FILMS**



(57) Abstract: A low dielectric constant hydrogenated silicon-oxycarbide (SiCO:H) film is prepared by conducting chemical vapor deposition using, together with an O<sub>2</sub>-containing gas plasma, an organosilicon or organosilicate compound having at least one vinyl or ethynyl group, or a mixture of a saturated organosilicon or organosilicate compound and an unsaturated hydrocarbon.

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## METHOD FOR PREPARING LOW DIELECTRIC FILMS

### FIELD OF THE INVENTION

5       The present invention relates to an improved plasma chemical vapor deposition (CVD) method for preparing a low dielectric constant hydrogenated silicon-oxycarbide (SiCO:H) film.

### BACKGROUND OF THE INVENTION

10

With ever decreasing size of electronic devices utilized in ULSI (ultra-large-scale integrated) circuits, there has emerged the problem of increased capacitance of intralayers and/or interlayers, causing signal delays.

Therefore, there has been a need to develop a low dielectric constant (k)  
15 material having a k-value lower than that of the conventional silicon dioxide (SiO<sub>2</sub>) or fluorinated silicon oxide (SiOF).

US Patent No. 6,147,009 discloses a low dielectric constant material produced by reacting the vapor of a precursor containing atoms of Si, C, O and H in a parallel plate plasma enhanced chemical vapor deposition chamber, the  
20 precursor being a molecule with a ring structure such as 1,3,5,7-tetramethylcyclotetrasiloxane(TMCTS,  $C_4H_{16}O_4Si_4$ ), tetraethylcyclotetrasiloxane( $C_8H_{24}O_4Si_4$ ) or decamethylcyclopentasiloxane( $C_{10}H_{30}O_5Si_5$ ), with or without added oxygen. However, the dielectric constant of the disclosed film is still high, in the range  
25 of 3.3 to 4.0. To further reduce the dielectric constant of the material described in the patent, US Patent No. 6,312,793 proposes a low k material consisting of two or more phases. However, the multi-phase material still has a k-value of more than 3.2.

Accordingly, the present inventors have endeavored to develop a novel  
30 material having a dielectric constant lower than those of conventional materials.

### SUMMARY OF THE INVENTION

35       It is, therefore, an object of the present invention to provide a method

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for preparing an improved low dielectric constant hydrogenated silicon-oxycarbide (SiCO:H) film using a chemical vapor deposition (CVD) technique.

It is another object of the present invention to provide a SiCOH film  
5 having a dielectric constant (k) lower than those of conventional low dielectric materials.

In accordance with one aspect of the present invention, there is provided a method for preparing a low dielectric constant hydrogenated silicon-oxycarbide (SiCO:H) film which comprises conducting chemical vapor  
10 deposition using, together with an O<sub>2</sub>-containing gas plasma, an organosilicon or organosilicate compound having at least one vinyl or ethynyl group, or a mixture of a saturated organosilicon or organosilicate compound and an unsaturated hydrocarbon.

In accordance with another aspect of the present invention, there is  
15 provided a low dielectric constant SiCOH thin film having a dielectric constant (k) of 2.6 or below, prepared by said method.

#### BRIEF DESCRIPTION OF THE DRAWINGS

20 The above and other objects and features of the present invention will become apparent from the following description thereof, when taken in conjunction with the accompanying drawings which respectively show:

FIGS. 1a and 1b: schematic diagrams of the plasma reactor used in forming a thin film in accordance with the present invention;

25 FIG. 2: variations in carbon contents of the film obtained in Example 1 of the present invention with O<sub>2</sub>/VTMS flow ratio, respectively;

FIG. 3: variations in dielectric constants of the films obtained in Example 1 and Comparative Example 1 of the present invention with O<sub>2</sub>/VTMS and O<sub>2</sub>/4MS flow ratio, respectively;

30 FIGS. 4 and 5: changes in the dielectric constant of the film obtained in Example 1 as function of annealing temperature and annealing time, respectively;

FIGS. 6 and 7: variations in carbon contents and dielectric constants of the film obtained in Example 2 of the present invention with O<sub>2</sub>/(4MS+C<sub>2</sub>F<sub>4</sub>)  
35 flow ratio, respectively;

FIGS. 8 and 9: changes in the dielectric constant of the film obtained in

Example 2 as function of annealing temperature and annealing time, respectively;

FIG. 10: variations in carbon contents of the film obtained in Example 3 of the present invention with  $O_2$ /TVTMCTSO flow ratio, respectively;

5 FIG. 11: variations in dielectric constants of the films obtained in Example 3 and Comparative Example 2 of the present invention with  $O_2$ /TVTMCTSO and  $O_2$ /TMCTSO flow ratio, respectively;

FIGS. 12 and 13: changes in the dielectric constant of the film obtained in Example 3 as function of annealing temperature and annealing time,  
10 respectively;

FIGS. 14 and 15: variations in carbon contents and dielectric constants of the film obtained in Example 4 of the present invention with  $O_2$ /(TMCTSO+C<sub>2</sub>H<sub>4</sub>) flow ratio, respectively;

FIGS. 16 and 17: changes in the dielectric constant of the film obtained  
15 in Example 4 as function of annealing temperature and annealing time, respectively;

FIGS. 18 and 19: variations in carbon contents and dielectric constants of the film obtained in Example 5 of the present invention with  $O_2$ /DADMS flow ratio, respectively;

20 FIGS. 20 and 21: changes in the dielectric constant of the film obtained in Example 5 as function of annealing temperature and annealing time, respectively;

FIGS. 22 and 23: variations in carbon contents and dielectric constants of the film obtained in Example 6 of the present invention with  $O_2$ /DVTMDSO  
25 flow ratio, respectively;

FIGS. 24 and 25: changes in the dielectric constant of the film obtained in Example 6 as function of annealing temperature and annealing time, respectively;

FIGS. 26 and 27: variations in carbon contents and dielectric constants  
30 of the film obtained in Example 7 of the present invention with  $O_2$ /VTMOS flow ratio, respectively;

FIGS. 28 and 29: changes in the dielectric constant of the film obtained in Example 7 as function of annealing temperature and annealing time, respectively;

35 FIGS. 30 and 31: variations in carbon contents and dielectric constants of the film obtained in Example 8 of the present invention with  $O_2$ /ETMS flow

ratio, respectively;

FIGS. 32 and 33: changes in the dielectric constant of the film obtained in Example 8 as function of annealing temperature and annealing time, respectively;

5 FIGS. 34 and 35: variations in carbon contents and dielectric constants of the films obtained in Example 9 of the present invention with  $O_2/(HMDSO+C_2H_4)$  flow ratio, respectively; and

FIGS. 36 and 37: changes in the dielectric constant of the film obtained in Example 9 as function of annealing temperature and annealing time,  
10 respectively.

### DETAILED DESCRIPTION OF THE INVENTION

The present invention provides a method for preparing a low dielectric  
15 constant hydrogenated silicon-oxycarbide ( $SiCO:H$ ) film by way of conducting chemical vapor deposition using, together with an  $O_2$ -containing gas plasma, an organosilicon or organosilicate compound having at least one vinyl or ethinyl group, or a mixture of a saturated organosilicon or organosilicate compound and an unsaturated hydrocarbon.

20 In accordance with the present invention, the process for forming a low dielectric constant  $SiCOH$  film may be conducted using a plasma CVD apparatus, e.g., a remote plasma CVD or a direct plasma CVD apparatus.

The remote plasma CVD apparatus shown in FIG. 1a comprises a quartz tube(6), an antenna(9), a matching box(5), a high-frequency electric  
25 power source(10), a mass flow controller(7) for feeding a silicon precursor, a precursor container(2), and a mass flow controller(8) for feeding a reactive gas. The antenna is wound around the outer periphery of the quartz tube(6), to thereby connect the antenna(9) and the matching box(5), which is connected to the high-frequency electric power source(10). The quartz  
30 tube(6) is connected to the mass flow controller(8) for feeding an  $O_2$ -containing gas via a tube. In the remote plasma CVD apparatus, the  $O_2$ -containing gas and the silicon precursor are fed to the matching box(5) separately, the precursor being led through a diffusion ring(3). Further, in the direct plasma CVD apparatus shown in FIG. 1b, the silicon precursor fed from  
35 the precursor container(2) and the  $O_2$ -containing gas fed from a reactive gas container(1) are mixed, and then supplied to the matching box(5).

In accordance with a preferred embodiment of the present invention, there is provided a method for preparing a low dielectric constant SiCOH material, which comprises conducting chemical vapor deposition using an unsaturated organosilicon or organosilicate compound having at least one vinyl or ethynyl group and an O<sub>2</sub>-containing gas plasma.

Representative examples of the unsaturated organosilicon or organosilicate compound having at least one vinyl or ethynyl group include vinyltrimethylsilane, vinyltriethylsilane, vinyltrimethoxysilane, vinyltriethoxysilane, 1,3,5-trivinyl-1,3,5-trimethylcyclotrisiloxane, 1,3,5,7-tetravinyl-1,3,5,7-tetramethylcyclotetrasiloxane, 1,3-divinyltetramethyldisiloxane, hexavinylidisiloxane, allyldimethylsilane, allyldimethoxysilane, ethynyltrimethylsilane, ethynyltriethylsilane and a mixture thereof.

The unsaturated organosilicon or organosilicate compound may be generated in situ, using an organosilicon or organosilicate having one or more halogen substituents.

In another preferred embodiment of the present invention, there is provided a method for preparing a low dielectric constant SiCOH material, which comprises conducting chemical vapor deposition using a mixture of a saturated organosilicon or organosilicate compound and an unsaturated hydrocarbon with an O<sub>2</sub>-containing gas plasma. The mixing ratio of the saturated organosilicon or organosilicate compound and the unsaturated hydrocarbon is preferably in the range from 1:0.1 to 1:10. If the mixing ratio is less than 0.1, the dielectric constant of the film becomes too high, while the physical properties of the film become unsatisfactory if the ratio is above 10.

In the above embodiment, the saturated organosilicon or organosilicate compound, or the unsaturated hydrocarbon may have one or more halogen substituents.

Representative examples of the saturated organosilicon or organosilicate compound include trimethylsilane, triethylsilane, trimethoxysilane, triethoxysilane, tetramethylsilane, tetraethylsilane, tetramethoxysilane, tetraethoxysilane, hexamethylcyclotrisiloxane, tetramethylcyclotetrasiloxane, tetraethylcyclotetrasiloxane, octamethylcyclotetrasiloxane, hexamethyldisiloxane, bistrimethylsilylmethane and a mixture thereof.

Representative examples of the unsaturated hydrocarbon are H<sub>2</sub>C=CH<sub>2</sub>, F<sub>2</sub>C=CF<sub>2</sub>, H<sub>2</sub>C=CF<sub>2</sub>, HFC=CFH, F<sub>2</sub>C=C=CF<sub>2</sub>, H<sub>2</sub>C=C=CF<sub>2</sub>, HFC=C=CFH,



HC CH, FC CH, FC CF,  $\text{Cl}_2\text{C}=\text{CCl}_2$ ,  $\text{H}_2\text{C}=\text{CCl}_2$ ,  $\text{HCIC}=\text{CCIH}$ ,  
 $\text{Cl}_2\text{C}=\text{C}=\text{CCl}_2$ ,  $\text{H}_2\text{C}=\text{C}=\text{CCl}_2$ ,  $\text{HCIC}=\text{C}=\text{CCIH}$ ,  $\text{ClC CH}$ ,  $\text{ClC CCl}$ ,  
 $\text{Br}_2\text{C}=\text{CBr}_2$ ,  $\text{H}_2\text{C}=\text{CBr}_2$ ,  $\text{HBrC}=\text{CBrH}$ ,  $\text{Br}_2\text{C}=\text{C}=\text{CBr}_2$ ,  $\text{H}_2\text{C}=\text{C}=\text{CBr}_2$ ,  
 $\text{HBrC}=\text{C}=\text{CBrH}$ ,  $\text{BrC CH}$ ,  $\text{BrC CBr}$ ,  $\text{I}_2\text{C}=\text{CI}_2$ ,  $\text{H}_2\text{C}=\text{CI}_2$ ,  $\text{HIC}=\text{CIH}$ ,  
 5  $\text{I}_2\text{C}=\text{C}=\text{CI}_2$ ,  $\text{H}_2\text{C}=\text{C}=\text{CI}_2$ ,  $\text{HIC}=\text{C}=\text{CIH}$ ,  $\text{IC CH}$  and  $\text{IC Cl}$ ; and preferred is  
 $\text{H}_2\text{C}=\text{CH}_2$  or  $\text{F}_2\text{C}=\text{CF}_2$ .

The  $\text{O}_2$ -containing gas which may be used in the present invention is selected from the group consisting of  $\text{O}_2$ ,  $\text{N}_2\text{O}$ ,  $\text{O}_3$ ,  $\text{H}_2\text{O}_2$ ,  $\text{CO}_2$ ,  $\text{H}_2\text{O}$  and a mixture thereof.

10 The method of the present invention may further comprise the step of annealing the deposited film, which may be carried out at a temperature ranging from 100 to 800°C for a period ranging from 0.5 to 8 hrs, preferably at 450°C for 1 hr, to obtain a thermally stable low dielectric constant SiCOH film.

The annealing step may also include a rapid-thermal processing, which may  
 15 be conducted at a temperature ranging from 100 to 900°C for about 1 minute and a spike-heating step performed for 10 seconds.

Such low dielectric constant SiCOH material prepared in accordance with the method of the present invention has a dielectric constant (k) of 2.8 or below; and, further, the thermally stable SiCOH film formed after annealing  
 20 has an exceptionally low dielectric constant (k) in the range of 1.6 to 2.6, the dielectric constant (k) being controllable by adjusting the process variables.

The present invention is further described and illustrated in Examples provided below, which are, however, not intended to limit the scope of the present invention.

25

### Example 1

A SiCOH film was deposited on a Pt substrate using vinyltrimethylsilane(VTMS,  $\text{SiC}_3\text{H}_{12}$ ) and  $\text{O}_2$  in the direct plasma apparatus shown in FIG. 1b. The flow ratio of  $\text{O}_2$ /VTMS was varied in the range of 1  
 30 to 13.3 during the film deposition. The pressure and temperature in the reactor were 1mmHg and 30°C, respectively, and the applied plasma power, 60W. The film so deposited was annealed under an Ar atmosphere at a temperature in the range of 300 to 500°C, to obtain a low dielectric constant film.

35 As shown in FIG. 2, the respective carbon contents of the deposited film and the film annealed at 450°C become lower with the increasing flow

ratio of  $O_2/VTMS$ . FIG. 3 exhibits that the annealed film at  $450^\circ C$  has a dielectric constant ranging from 1.8 to 2.4, while the deposited film without the annealing has a dielectric constant ranging from 2.3 to 2.8. FIGS. 4 and 5 show the changes in the dielectric constant of the film obtained in Example 1 with the changes in the annealing temperature and annealing time at the  $O_2/VTMS$  flow ratio of 2, respectively.

### Comparative Example 1

The procedure of Example 1 was repeated using tetramethylsilane(4MS,  $SiC_4H_{12}$ ) in place of VTMS, to obtain a deposited film. As shown in FIG. 3, the film thus obtained has a dielectric constant ranging from 3.0 to 3.5, which is higher than that of the deposited film obtained in Example 1.

### Example 2

Except that a mixture of tetramethylsilane(4MS,  $SiC_4H_{12}$ ) and  $C_2F_4$  (1:1) was used instead of VTMS, the procedure of Example 1 was repeated to obtain a deposited film, which was subsequently annealed.

FIG. 6 and FIG. 7 show the carbon contents and dielectric constants of the deposited film and the annealed film, respectively. The deposited film has a dielectric constant of 3.0 or below, and the film annealed at  $450^\circ C$  has a dielectric constant of 2.5 or below. FIG. 8 and FIG. 9 show the effects of the annealing temperature (annealing time=0.5hr) and annealing time (annealing temperature= $400^\circ C$ ) at the  $O_2/(4MS+C_2F_4)$  flow ratio of 4 on the dielectric constant of the film, respectively. The film annealed at 300 to  $500^\circ C$  for 0.5 hr has a dielectric constant of 2.75 or below.

### Example 3

The procedure of Example 1 was repeated using tetravinyltetramethylcyclotetrasiloxane(TVTMCTSO,  $Si_4O_4C_{12}H_{24}$ ) in place of VTMS, to obtain a deposited film, which was subsequently annealed.

FIG. 10 and FIG. 11 show the carbon contents and dielectric constants of the deposited film and the annealed film, respectively. The deposited film has a dielectric constant of 2.4 or below, and the film annealed at  $450^\circ C$ , a dielectric constant of 2.2 or below. FIG. 12 and FIG. 13 show the effects of the annealing temperature (annealing time=0.5hr) and annealing time (annealing temperature= $450^\circ C$ ) at the  $O_2/TVTMCTSO$  flow ratio of 4 on the

dielectric constant of the film, respectively. The film annealed at 300 to 500°C for 0.5 hr has a dielectric constant of 2.1 or below.

### Comparative Example 2

5 The procedure of Example 1 was repeated using tetramethylcyclotetrasiloxane(TMCTSO,  $\text{Si}_4\text{O}_4\text{C}_4\text{H}_{16}$ ) in place of VTMS, to obtain a deposited film. As shown in FIG. 11, the film thus obtained had a dielectric constant ranging from 2.5 to 3.3, which is higher than that of the deposited film obtained in Example 3.

10

### Example 4

Except that a mixture of tetramethylcyclotetrasiloxane(TMCTSO,  $\text{Si}_4\text{O}_4\text{C}_4\text{H}_{16}$ ) and  $\text{C}_2\text{H}_4$  (1:1) was used instead of VTMS, the procedure of Example 1 was repeated to obtain a deposited film, which was subsequently  
15 annealed.

FIG. 14 and FIG. 15 depict the carbon contents and dielectric constants of the deposited film and the annealed film, respectively. The deposited film has a dielectric constant of 2.3 or below, and the film annealed at 450°C has a dielectric constant of 2.2 or below. FIG. 16 and FIG. 17 show the  
20 effects of the annealing temperature (annealing time=0.5hr) and annealing time (annealing temperature=400°C) at the (TMCTSO+ $\text{C}_2\text{H}_4$ ) flow ratio of 2 on the dielectric constant of the film, respectively. The film annealed at 300 to 500°C for 0.5 hr has a dielectric constant of 2.05 or below.

### 25 Example 5

The procedure of Example 1 was repeated using diallyldimethylsilane (DADMS,  $\text{SiC}_8\text{H}_{16}$ ) in place of VTMS, to obtain a deposited film, which was subsequently annealed.

FIG. 18 and FIG. 19 show the carbon contents and dielectric constants  
30 of the deposited film and the annealed film, respectively. The deposited film has a dielectric constant of 2.8 or below, and the film annealed at 450°C, a dielectric constant of 2.4 or below. FIG. 20 and FIG. 21 show the effects of the annealing temperature (annealing time=0.5hr) and annealing time (annealing temperature=450°C) at the  $\text{O}_2$ /DADMS flow ratio of 4 on the  
35 dielectric constant of the film, respectively. The film annealed at 300 to 500°C for 0.5 hr has a dielectric constant of 2.35 or below.

### Example 6

Except that 1,3-divinyltetramethyldisiloxane(DVTMDSO,  $\text{Si}_2\text{OC}_8\text{H}_{18}$ ) was used in place of VTMS, the procedure of Example 1 was repeated to  
5 obtain a deposited film, which was subsequently annealed.

FIG. 22 and FIG. 23 show the carbon contents and dielectric constants of the deposited film and the annealed film, respectively. The deposited film has a dielectric constant of 2.9 or below, and the film annealed at 450°C has a dielectric constant of 2.4 or below. FIG. 24 and FIG. 25 show the  
10 effects of the annealing temperature (annealing time=0.5hr) and annealing time (annealing temperature=500°C) at the  $\text{O}_2$ /DVTMDSO flow ratio of 2 on the dielectric constant of the film, respectively. The film annealed at 300 to 500°C for 0.5 hr has a dielectric constant of 2.15 or below.

### 15 Example 7

The procedure of Example 1 was repeated using vinyltrimethoxysilane (VTMOS,  $\text{SiO}_3\text{C}_5\text{H}_{12}$ ) in place of VTMS, to obtain a deposited film, which was subsequently annealed.

FIG. 26 and FIG. 27 show the carbon contents and dielectric constants  
20 of the deposited film and the annealed film, respectively. The deposited film has a dielectric constant in the range of 2.2 to 2.75, and the film annealed at 450°C has a dielectric constant in the range of 1.9 to 2.55. FIG. 28 and FIG. 29 show the effects of the annealing temperature (annealing time=0.5hr) and annealing time (annealing temperature=450°C) at the  
25  $\text{O}_2$ /VTMOS flow ratio of 2 on the dielectric constant of the film, respectively.

The film annealed at 300 to 500°C for 0.5 hr has a dielectric constant of 2.4 or below.

### Example 8

30 Except that ethynyltrimethylsilane (ETMS,  $\text{SiC}_5\text{H}_{10}$ ) was used in place of VTMS, the procedure of Example 1 was repeated to obtain a deposited film, which was subsequently annealed.

FIG. 30 and FIG. 31 show the carbon contents and dielectric constants of the deposited film and the annealed film, respectively. The deposited  
35 film has a dielectric constant of 2.65 or below, and the film annealed at 450°C, a dielectric constant of 2.35 or below. FIG. 32 and FIG. 33 show

the effects of the annealing temperature (annealing time=0.5hr) and annealing time (annealing temperature=350°C) at the O<sub>2</sub>/ETMS flow ratio of 4 on the dielectric constant of the film, respectively. The film annealed at 300 to 500°C for 0.5 hr has a dielectric constant of 2.35 or below.

5

### Example 9

The procedure of Example 1 was repeated using a mixture of hexamethyldisiloxane(HMDSO, Si<sub>2</sub>OC<sub>6</sub>H<sub>18</sub>) and C<sub>2</sub>H<sub>4</sub> (1:2) in place of VTMS, to obtain a deposited film, which was subsequently annealed.

10 FIG. 34 and FIG. 35 show the carbon contents and dielectric constants of the deposited film and the annealed film, respectively. The deposited film has a dielectric constant of 3.0 or below, and the film annealed at 450°C, a dielectric constant of 2.05 or below. FIG. 36 and FIG. 37 show the effects of the annealing temperature (annealing time=0.5hr) and annealing  
15 time (annealing temperature=450°C) at the O<sub>2</sub>/(HMDSO+C<sub>2</sub>H<sub>4</sub>) flow ratio of 4 on the dielectric constant of the film, respectively. The film annealed at 300 to 500°C for 0.5 hr has a dielectric constant ranging from 1.7 to 1.9.

As can be seen from the above results, the low dielectric constant SiCOH film prepared by conducting CVD using an O<sub>2</sub>-containing gas plasma  
20 and an unsaturated organosilicon or organosilicate compound, or a mixture of a saturated organosilicon or organosilicate compound and an unsaturated hydrocarbon in accordance with the present invention has a dielectric constant of 2.6 or below, which is exceptionally lower than conventional low k-materials.

25 While the subject invention have been described and illustrated with respect to the preferred embodiments only, various changes and modifications may be made therein without departing from the essential concept of the present invention which should be limited only by the scope of the appended claims.

What is claimed is:

1. A method for preparing a low dielectric constant hydrogenated silicon-oxycarbide (SiCO:H) film which comprises conducting chemical vapor deposition using, together with an O<sub>2</sub>-containing gas plasma, an organosilicon or organosilicate compound having at least one vinyl or ethynyl group, or a mixture of a saturated organosilicon or organosilicate compound and an unsaturated hydrocarbon.
2. The method of claim 1, wherein the mixture of a saturated organosilicon or organosilicate compound and an unsaturated hydrocarbon has a mixing ratio ranging from 1:0.1 to 1:10.
3. The method of claim 1, wherein the organosilicon or organosilicate compound having at least one vinyl or ethynyl group is selected from the group consisting of vinyltrimethylsilane, vinyltriethylsilane, vinyltrimethoxysilane, vinyltriethoxysilane, 1,3,5-trivinyl-1,3,5-trimethylcyclotrisiloxane, 1,3,5,7-tetravinyl-1,3,5,7-tetramethylcyclotetrasiloxane, 1,3-divinyltetramethyldisiloxane, hexavinylidisiloxane, allyldimethylsilane, allyldimethoxysilane, ethynyltrimethylsilane, ethynyltriethylsilane and a mixture thereof.
4. The method of claim 3, wherein the organosilicon or organosilicate compound having at least one vinyl or ethynyl group is selected from the group consisting of vinyltrimethylsilane, 1,3,5,7-tetravinyl-1,3,5,7-tetramethylcyclotetrasiloxane, allyldimethylsilane, 1,3-divinyltetramethyldisiloxane, vinyltrimethoxysilane and ethynyltrimethylsilane.
5. The method of claim 1, wherein the saturated organosilicon or organosilicate compound is selected from the group consisting of trimethylsilane, triethylsilane, trimethoxysilane, triethoxysilane, tetramethylsilane, tetraethylsilane, tetramethoxysilane, tetraethoxysilane, hexamethylcyclotrisiloxane, tetramethylcyclotetrasiloxane, tetraethylcyclotetrasiloxane, octamethylcyclotetrasiloxane, hexamethyldisiloxane, bistrimethylsilylmethane, vinyltrimethylsilane,

vinyltriethylsilane, vinyltrimethoxysilane, vinyltriethoxysilane,  
 1,3,5-trivinyl-1,3,5-trimethylcyclotrisiloxane,  
 1,3,5,7-tetravinyl-1,3,5,7-tetramethylcyclotetrasiloxane,  
 1,3-divinyldimethyltetramethyldisiloxane, hexavinylbisiloxane, allyldimethylsilane,  
 5 allyldimethoxysilane, ethynyltrimethylsilane, ethynyltriethylsilane and a  
 mixture thereof.

6. The method of claim 5, wherein the saturated organosilicon or  
 organosilicate compound is selected from the group consisting of  
 10 tetramethylsilane, hexamethyldisiloxane and tetramethylcyclotetrasiloxane.

7. The method of claim 1, wherein the unsaturated hydrocarbon is selected  
 from the group consisting of  $\text{H}_2\text{C}=\text{CH}_2$ ,  $\text{F}_2\text{C}=\text{CF}_2$ ,  $\text{H}_2\text{C}=\text{CF}_2$ ,  $\text{HFC}=\text{CFH}$ ,  
 $\text{F}_2\text{C}=\text{C}=\text{CF}_2$ ,  $\text{H}_2\text{C}=\text{C}=\text{CF}_2$ ,  $\text{HFC}=\text{C}=\text{CFH}$ ,  $\text{HC CH}$ ,  $\text{FC CH}$ ,  $\text{FC CF}$ ,  
 15  $\text{Cl}_2\text{C}=\text{CCl}_2$ ,  $\text{H}_2\text{C}=\text{CCl}_2$ ,  $\text{HClC}=\text{CClH}$ ,  $\text{Cl}_2\text{C}=\text{C}=\text{CCl}_2$ ,  $\text{H}_2\text{C}=\text{C}=\text{CCl}_2$ ,  
 $\text{HClC}=\text{C}=\text{CClH}$ ,  $\text{ClC CH}$ ,  $\text{ClC CCl}$ ,  $\text{Br}_2\text{C}=\text{CBr}_2$ ,  $\text{H}_2\text{C}=\text{CBr}_2$ ,  $\text{HBrC}=\text{CBrH}$ ,  
 $\text{Br}_2\text{C}=\text{C}=\text{CBr}_2$ ,  $\text{H}_2\text{C}=\text{C}=\text{CBr}_2$ ,  $\text{HBrC}=\text{C}=\text{CBrH}$ ,  $\text{BrC CH}$ ,  $\text{BrC CBr}$ ,  
 $\text{I}_2\text{C}=\text{CI}_2$ ,  $\text{H}_2\text{C}=\text{CI}_2$ ,  $\text{HIC}=\text{CIH}$ ,  $\text{I}_2\text{C}=\text{C}=\text{CI}_2$ ,  $\text{H}_2\text{C}=\text{C}=\text{CI}_2$ ,  $\text{HIC}=\text{C}=\text{CIH}$ ,  
 $\text{IC CH}$  and  $\text{IC Cl}$ .

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8. The method of claim 7, wherein the unsaturated hydrocarbon is  
 $\text{H}_2\text{C}=\text{CH}_2$  or  $\text{F}_2\text{C}=\text{CF}_2$ .

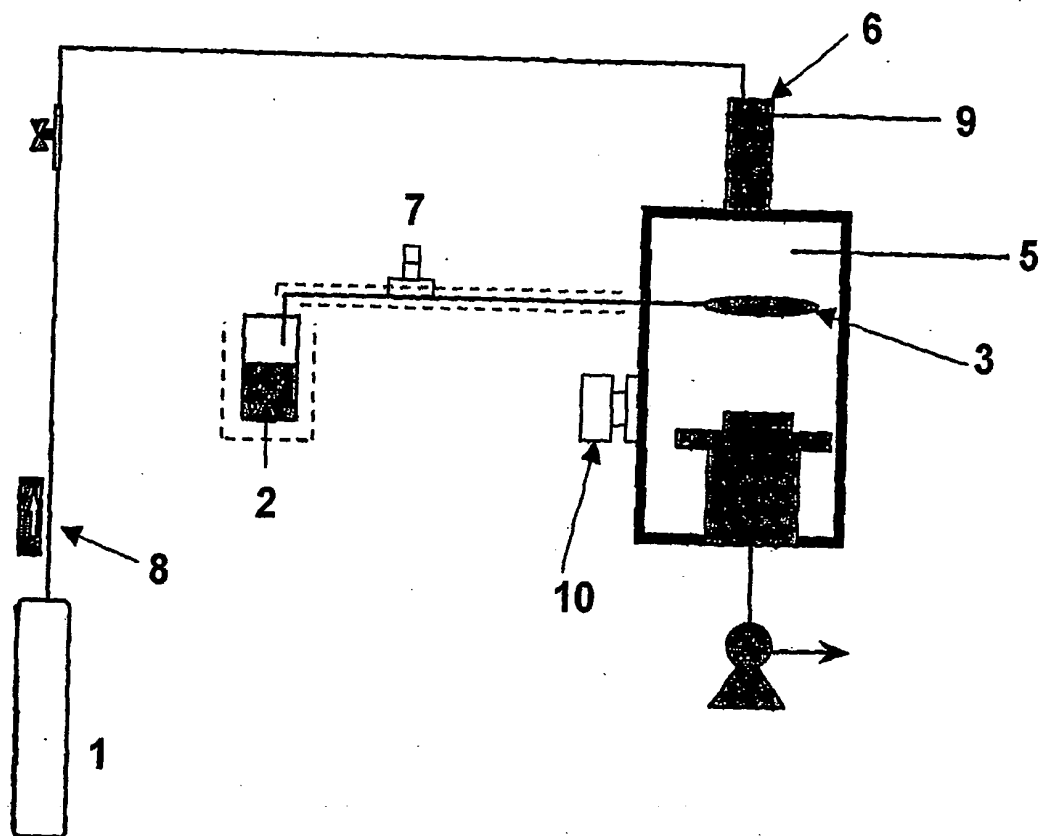
9. The method of claim 1, wherein the  $\text{O}_2$ -containing gas is selected from  
 25 the group consisting of  $\text{O}_2$ ,  $\text{N}_2\text{O}$ ,  $\text{O}_3$ ,  $\text{H}_2\text{O}_2$ ,  $\text{CO}_2$ ,  $\text{H}_2\text{O}$  and a mixture thereof.

10. The method of claim 1 further comprising the step of annealing the  
 deposited film at a temperature ranging from 100 to 500°C for 0.5 to 8 hrs.

30 11. A low dielectric constant hydrogenated silicon-oxycarbide ( $\text{SiCO:H}$ )  
 film prepared by the method of claim 1.

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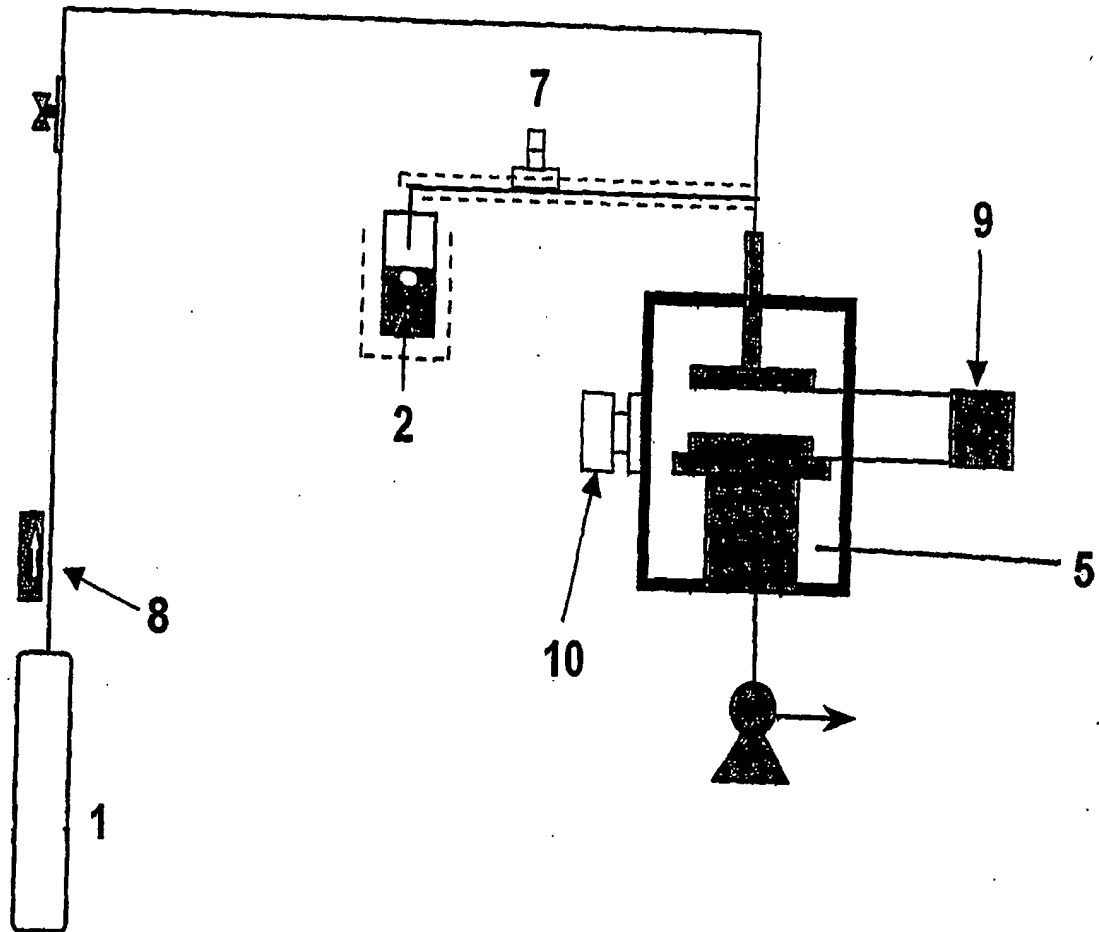
FIG. 1a





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FIG. 1b



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FIG. 2

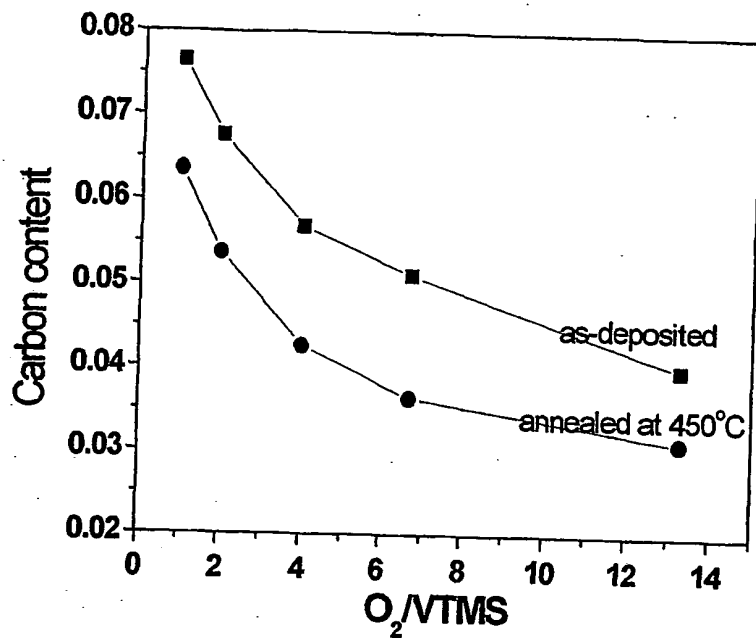
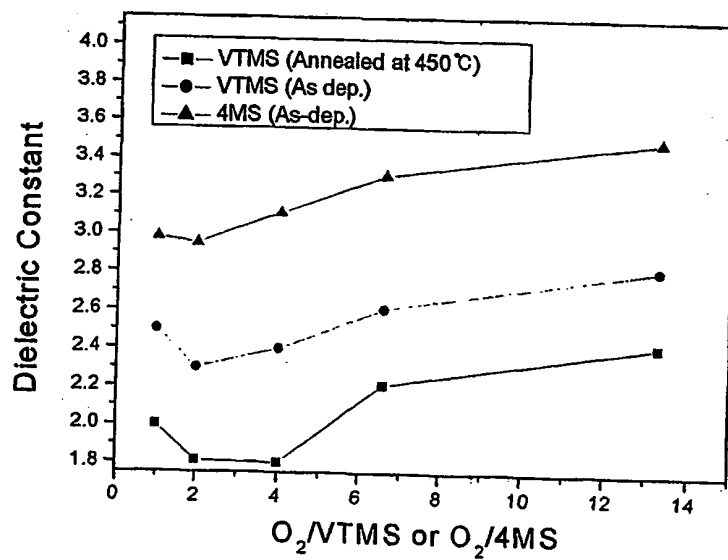


FIG. 3



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FIG. 4

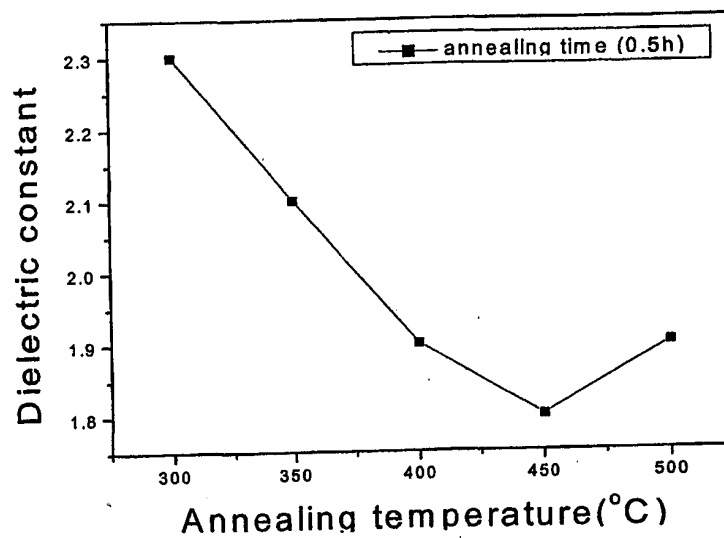
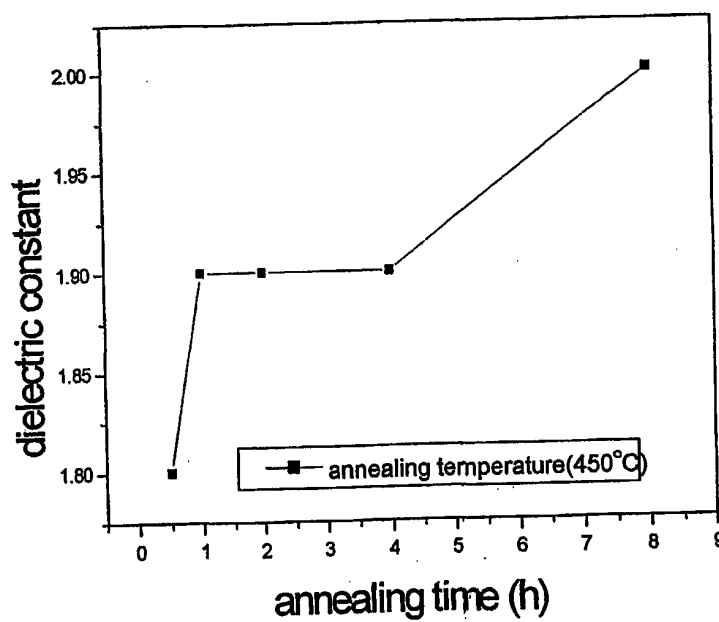


FIG. 5



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FIG. 6

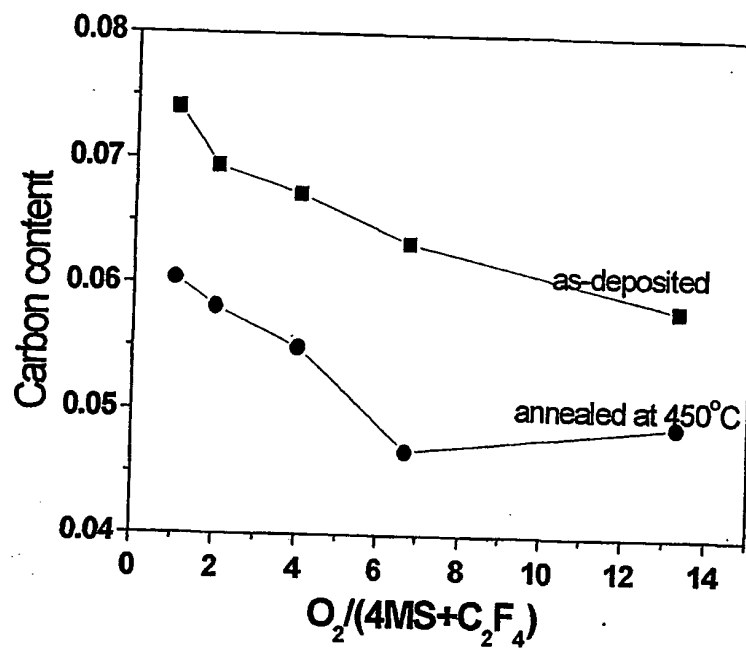
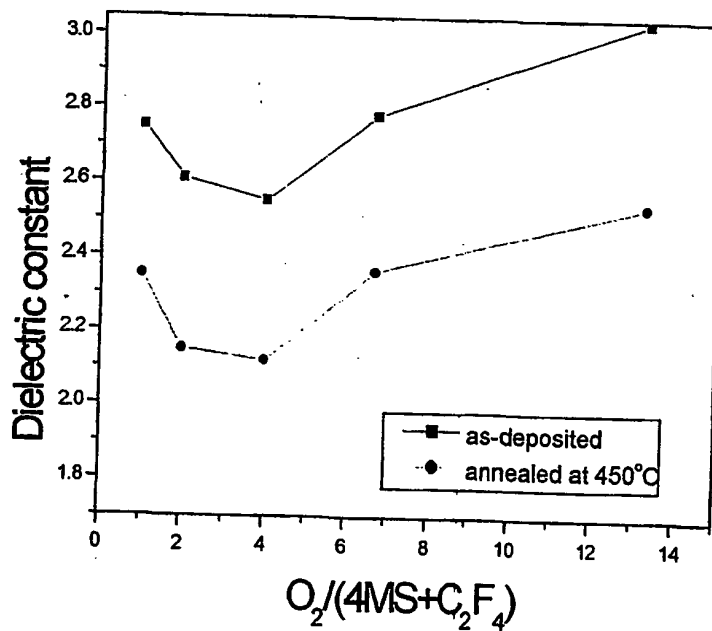


FIG. 7



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FIG. 8

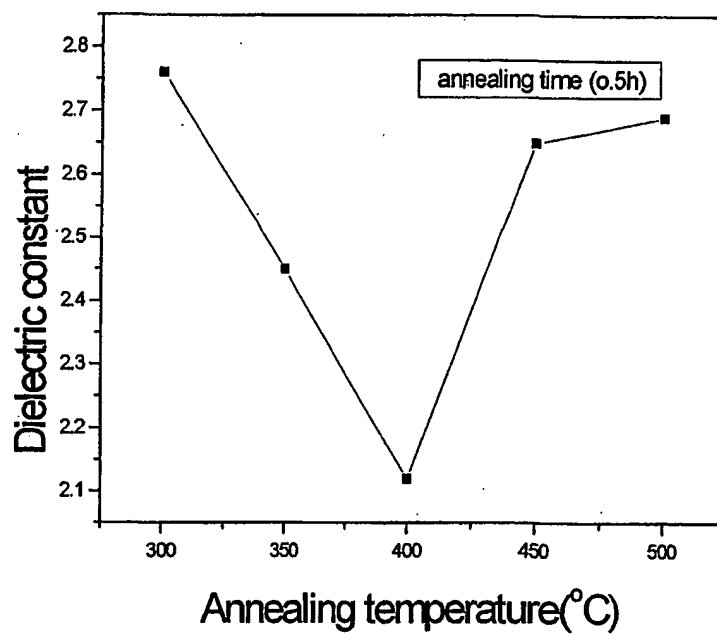
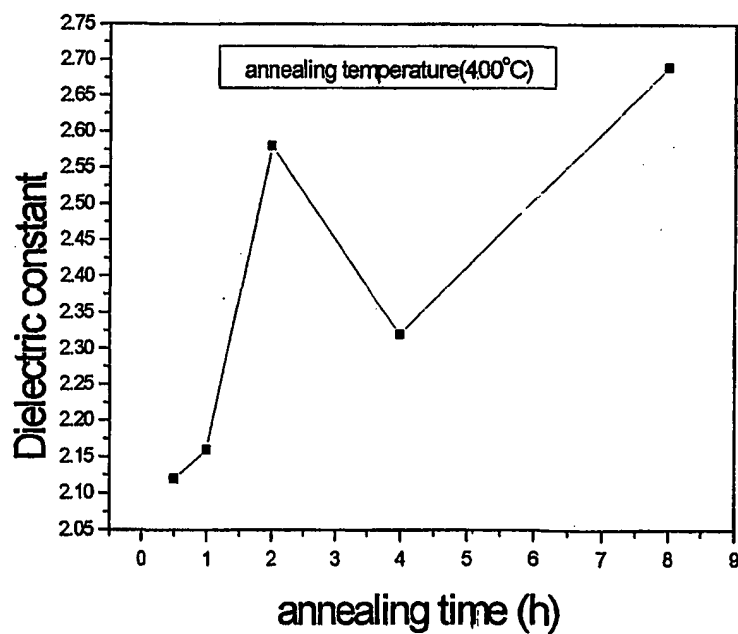


FIG. 9



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FIG. 10

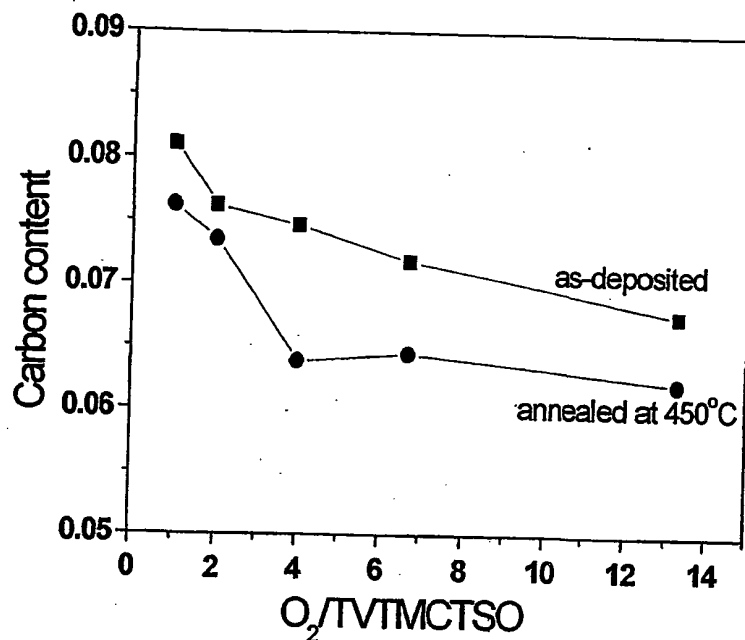
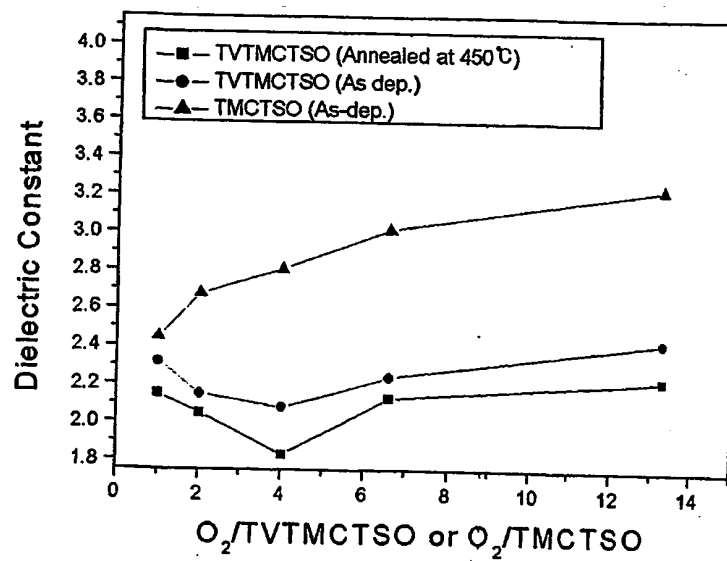


FIG. 11



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FIG. 12

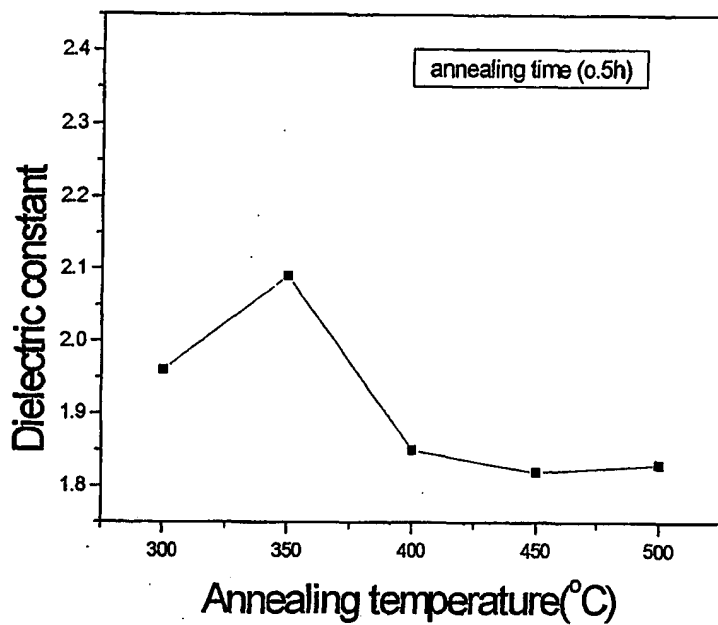
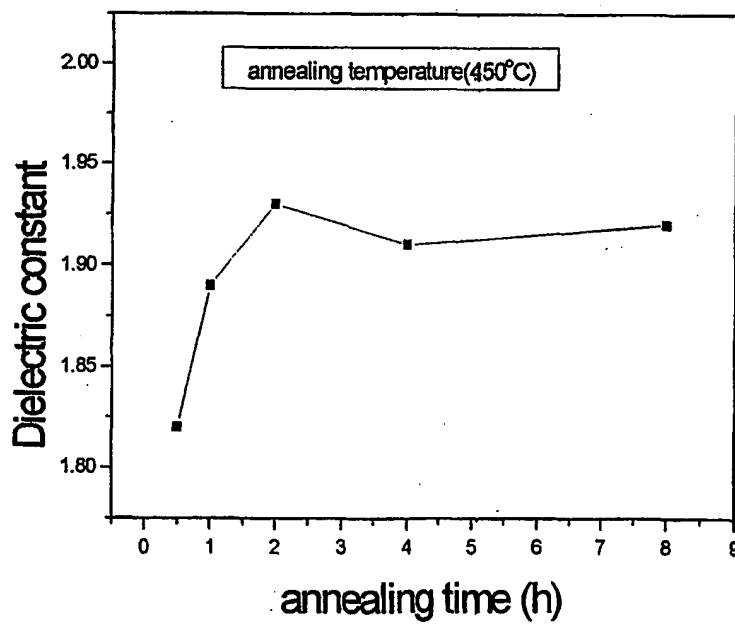


FIG. 13



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FIG. 14

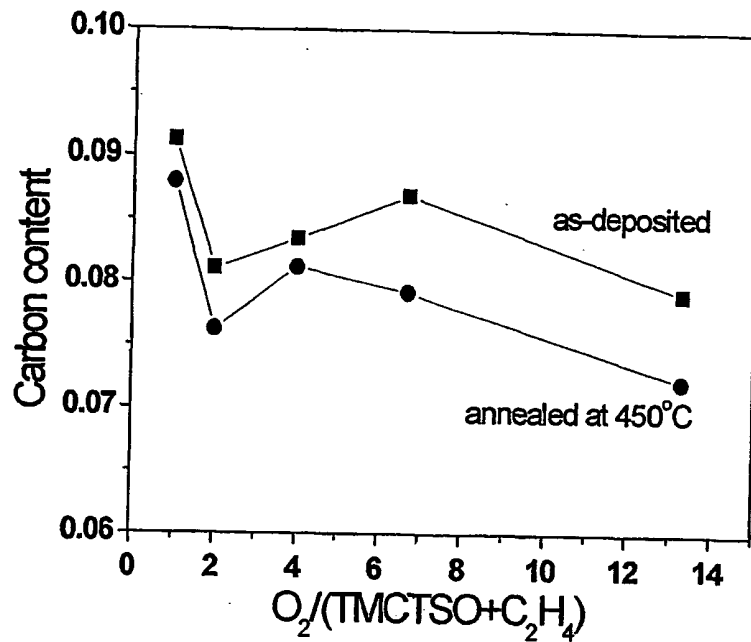
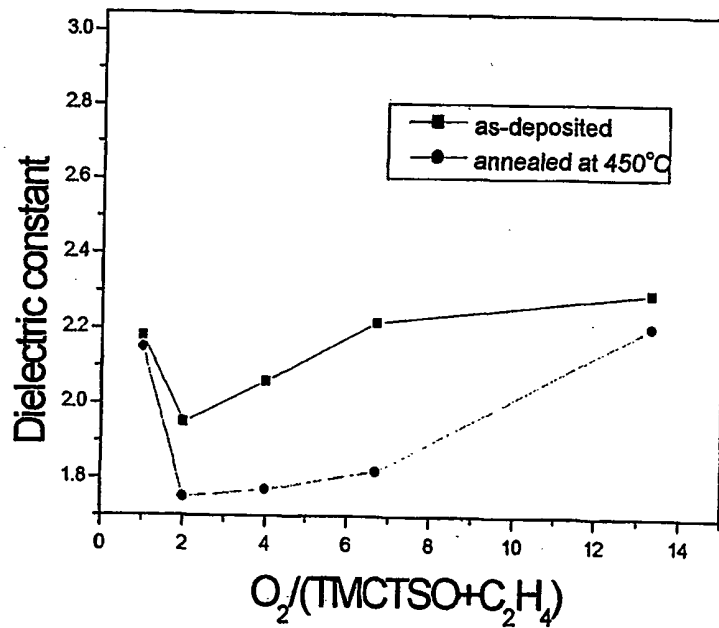
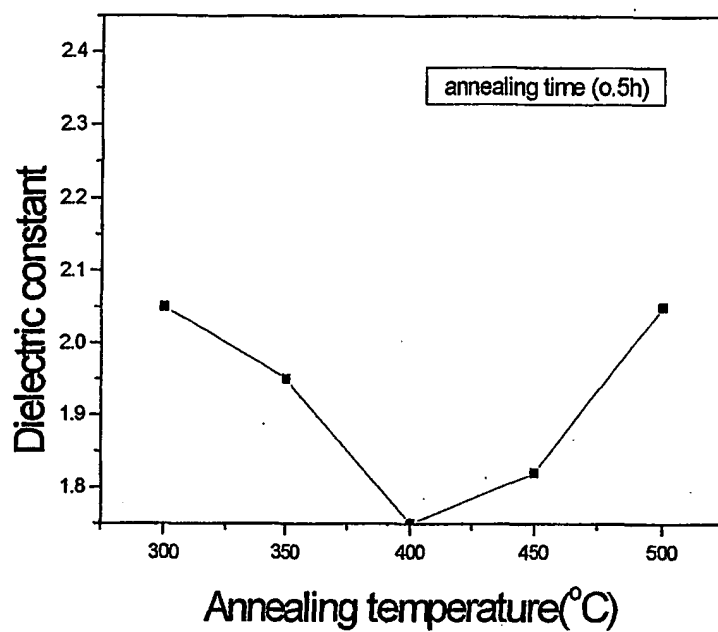
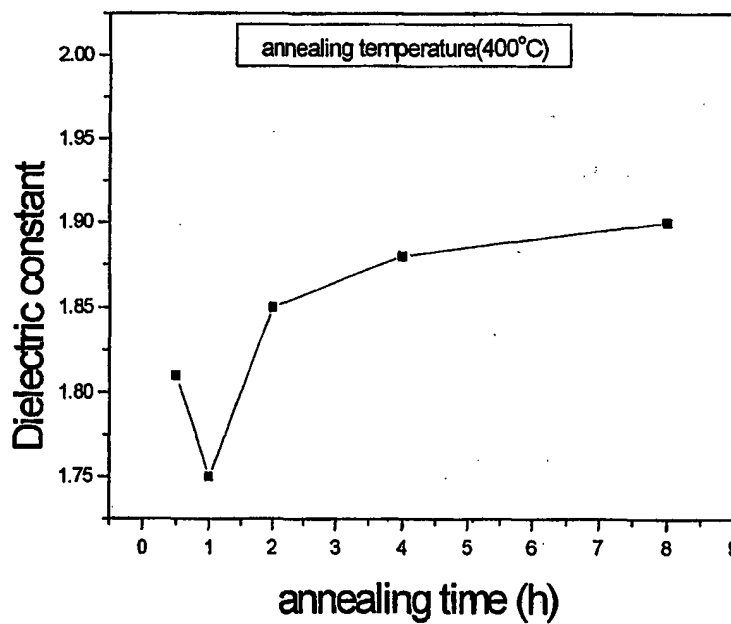


FIG. 15





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*FIG. 16**FIG. 17*

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FIG. 18

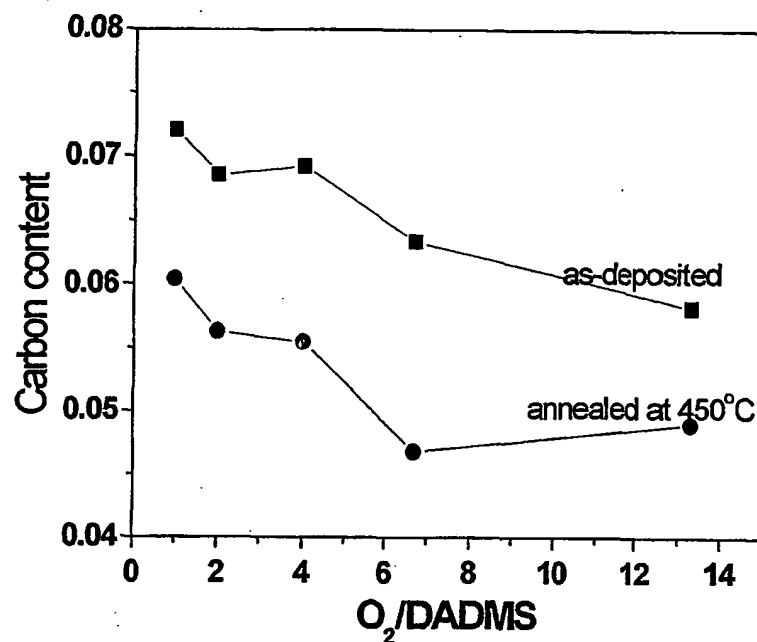
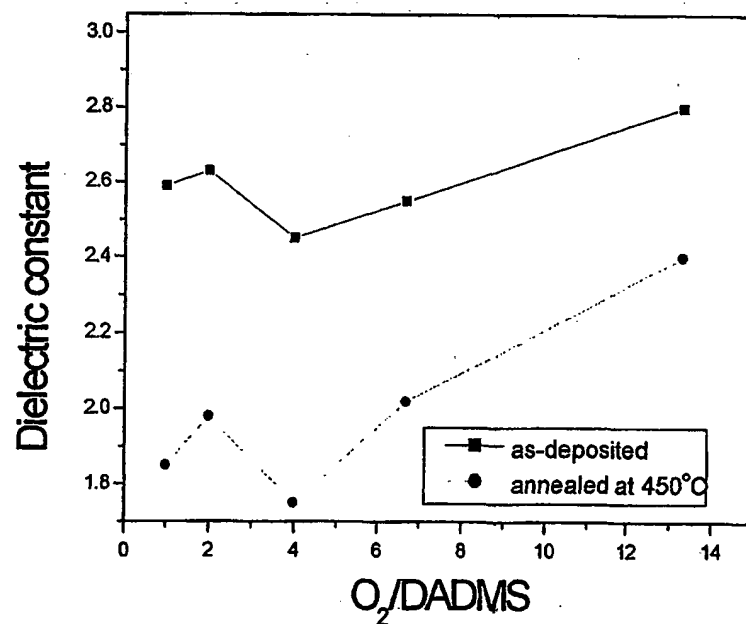
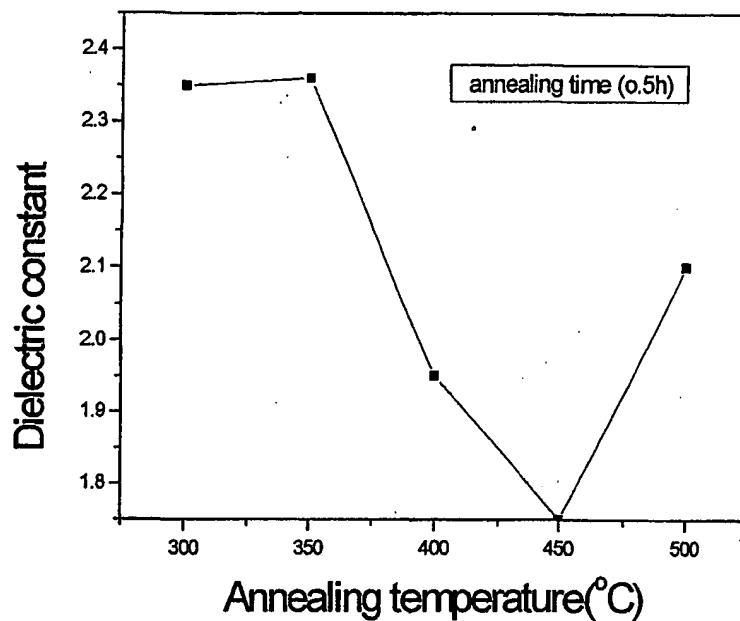
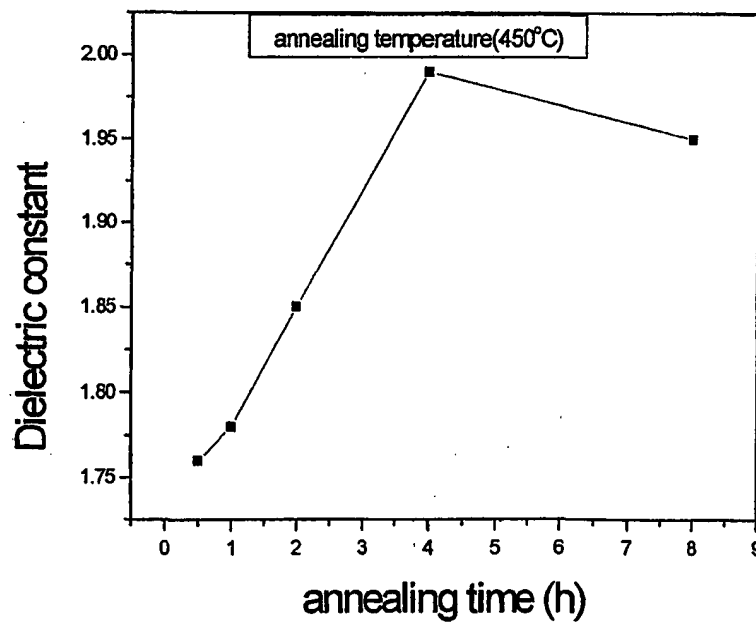


FIG. 19



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*FIG. 20**FIG. 21*

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FIG. 22

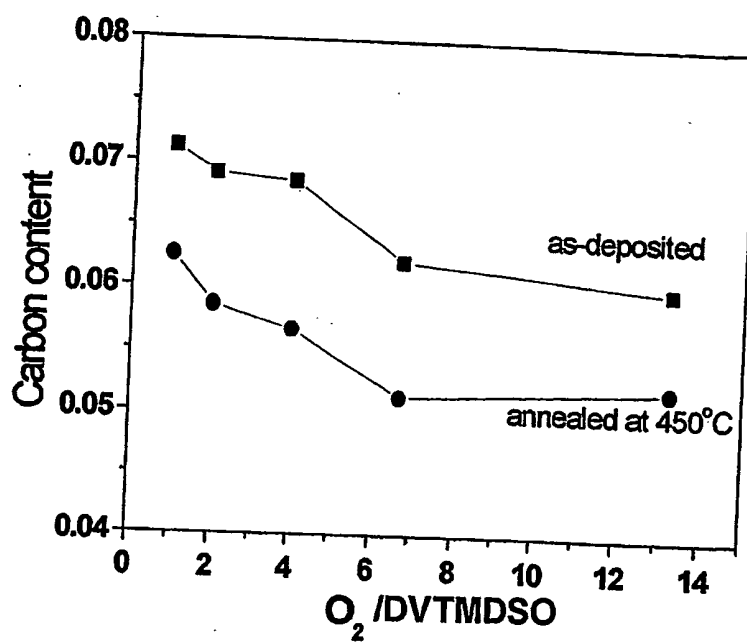
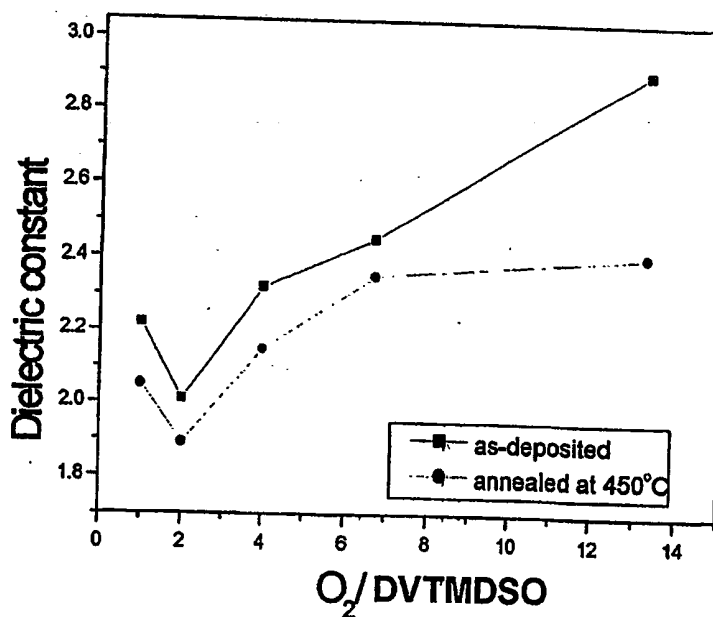


FIG. 23



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FIG. 24

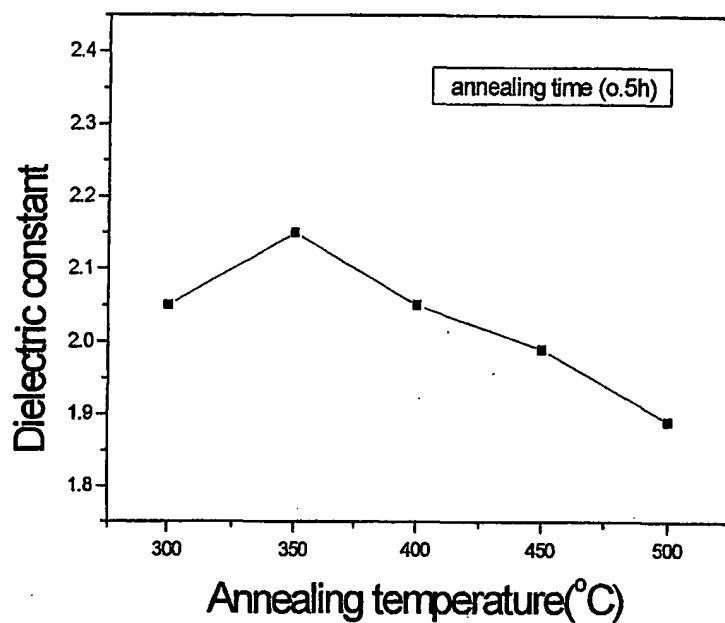
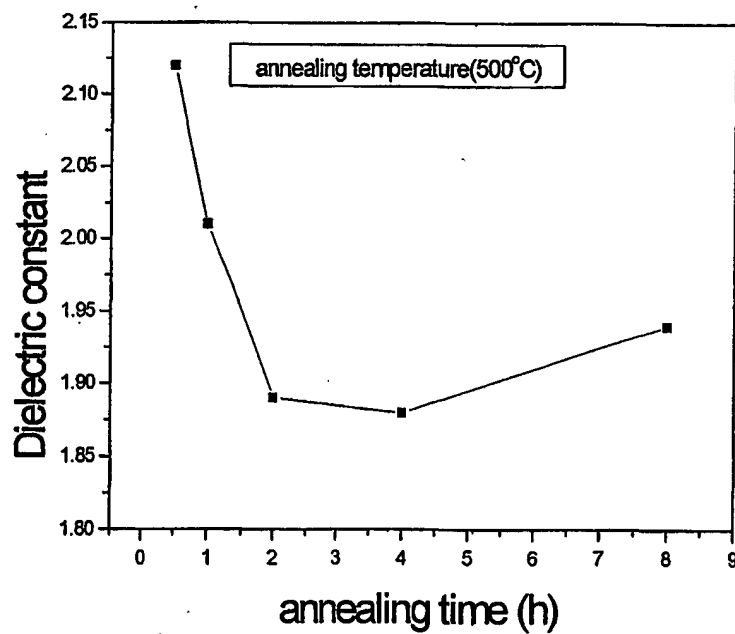


FIG. 25



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FIG. 26

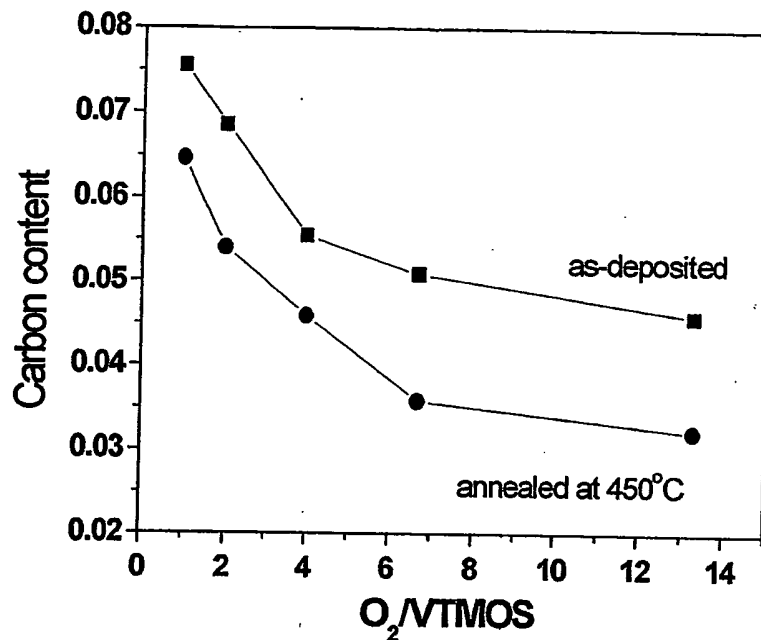
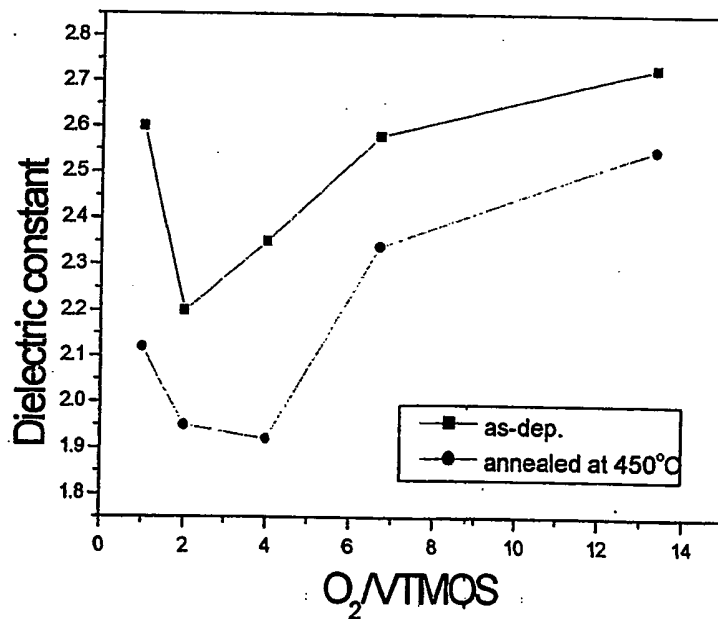
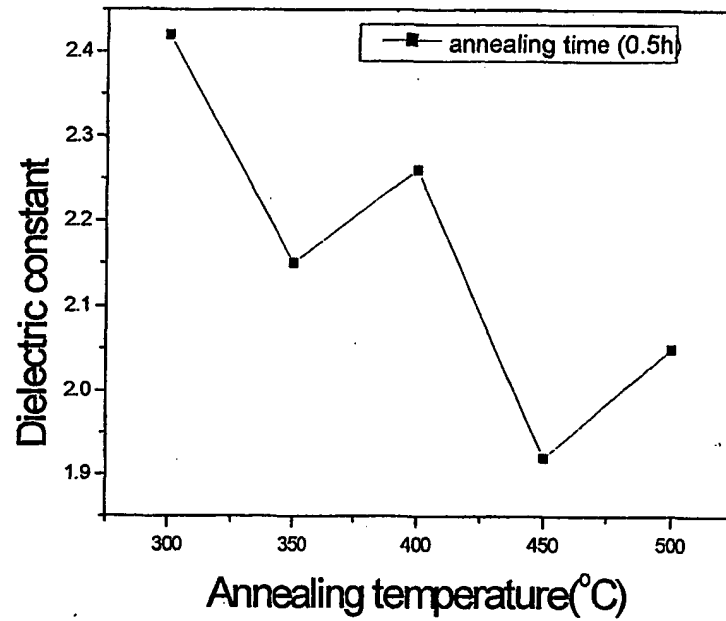
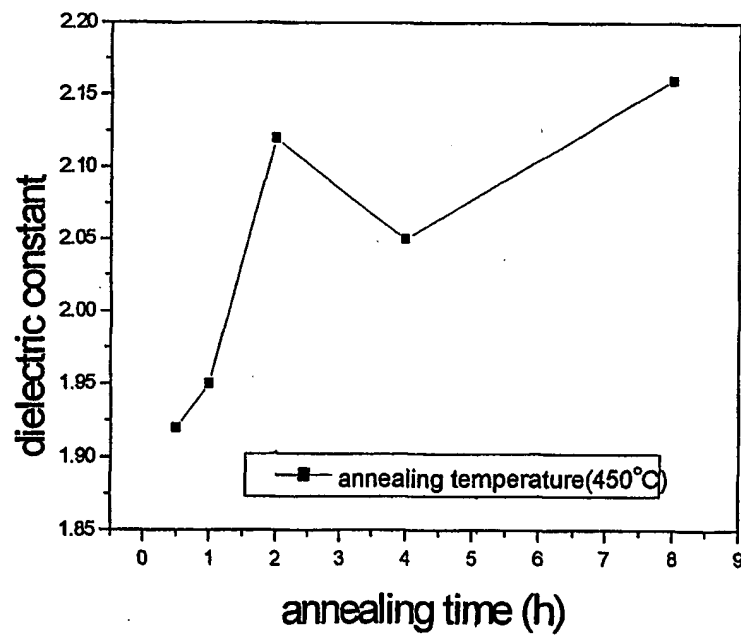


FIG. 27



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*FIG. 28**FIG. 29*

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FIG. 30

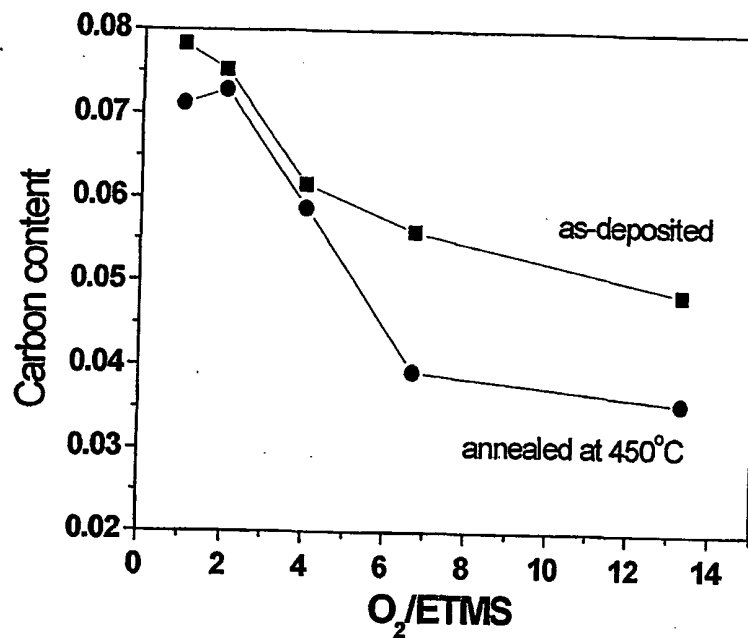
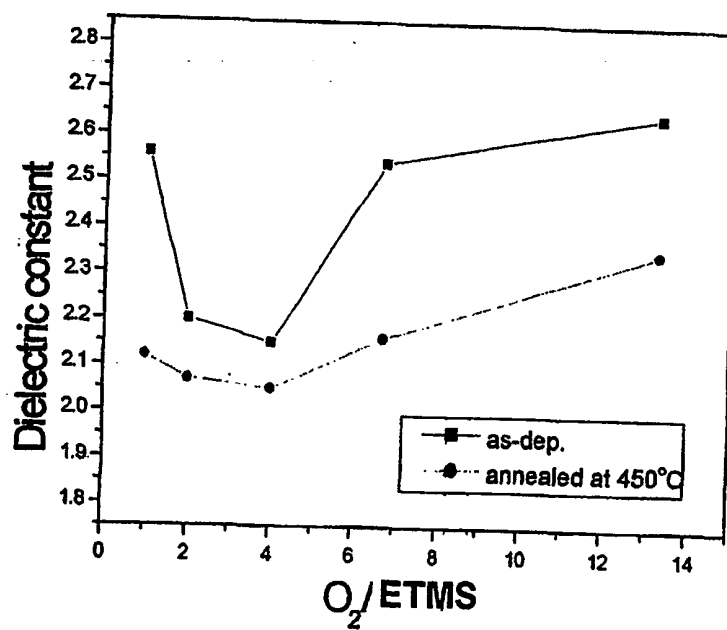
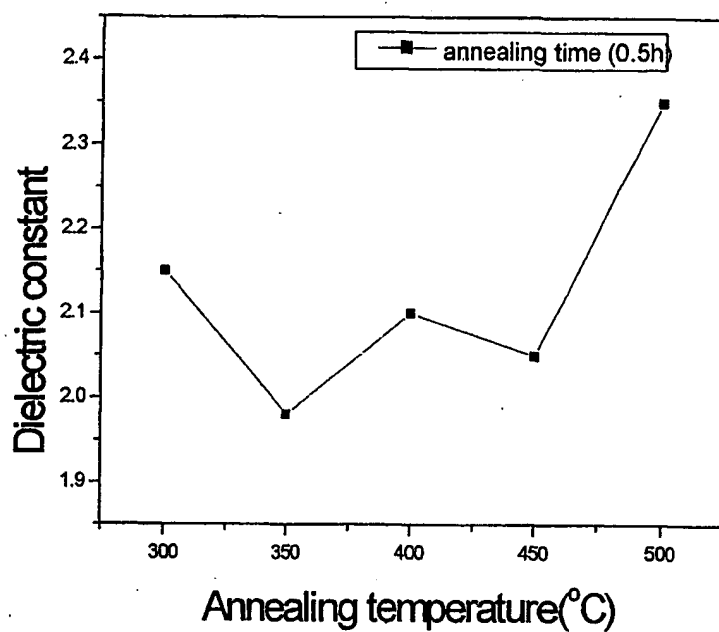
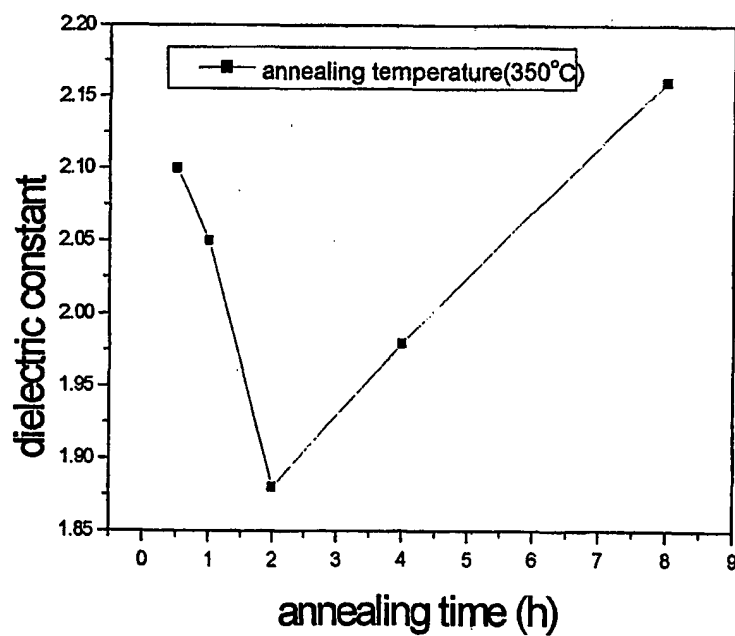


FIG. 31





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*FIG. 32**FIG. 33*

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FIG. 34

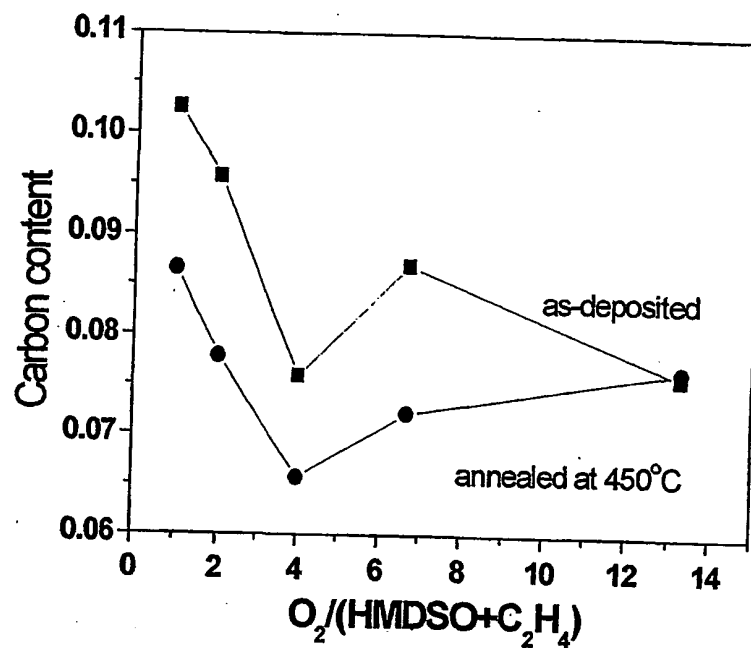
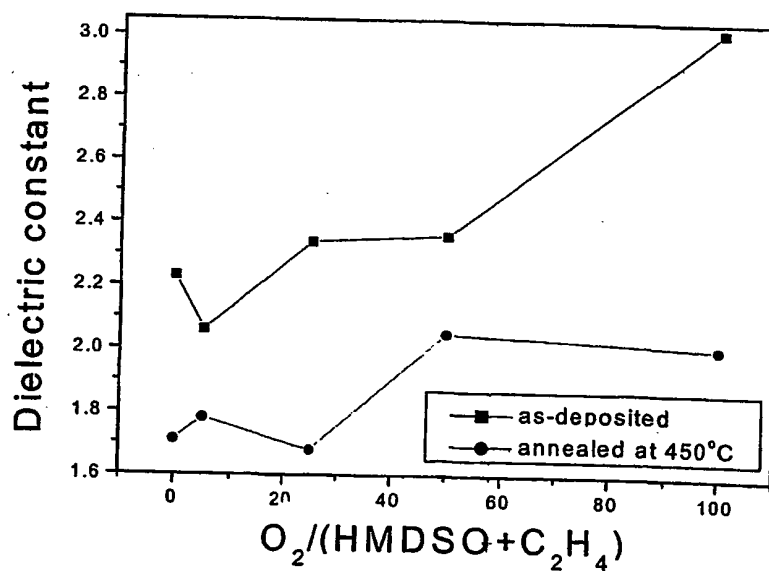
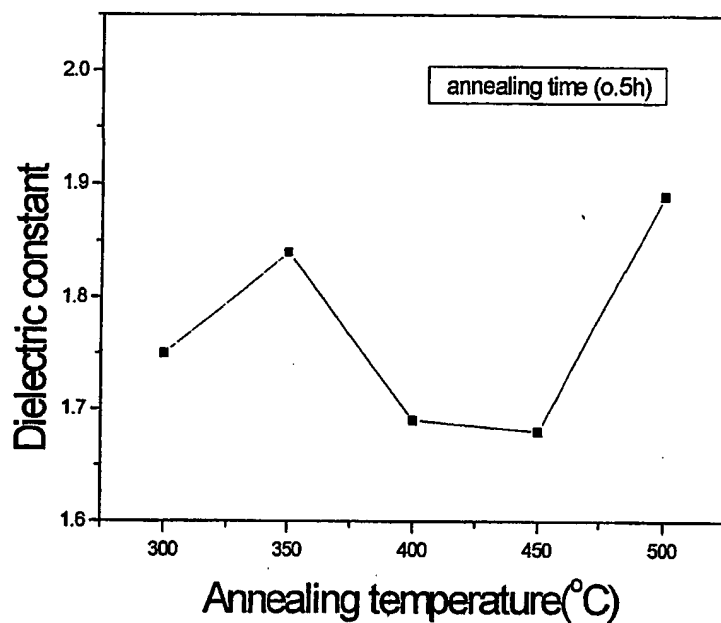
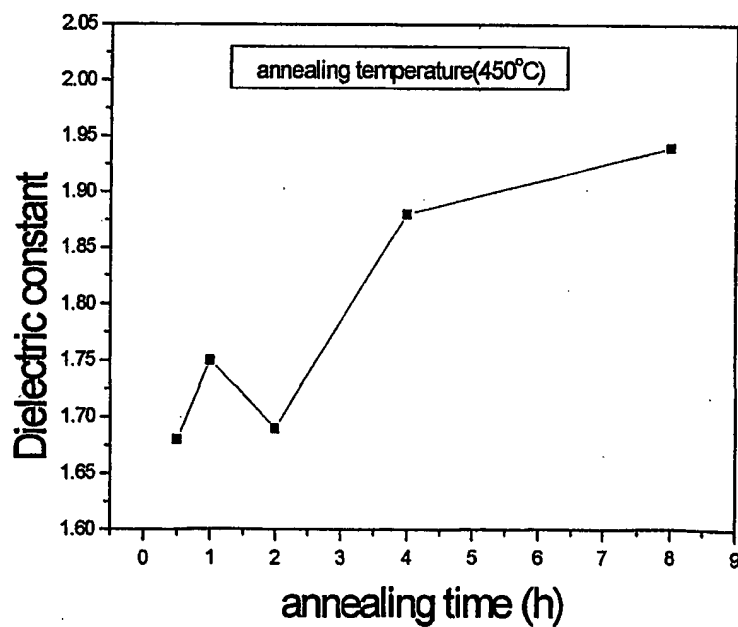


FIG. 35



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*FIG. 36**FIG. 37*

# INTERNATIONAL SEARCH REPORT

International application No.  
PCT/KR02/01238

## A. CLASSIFICATION OF SUBJECT MATTER

IPC7 H01L 21/22

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC7 C23C 16/32, C23C 16/30, B32B 3/00

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)  
USPAT, FPD, PAJ

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	EP, A, 0 960 958 (DOW CORNING CORP.) 01 December 1999 See page 2, line 28 - line 35, page 2, line 48 - page, 3, line 1	1, 3, 4, 5, 6, 11
Y	WO, A, 99/41423 (APPLIED MATERIALS INC.) 10 February 1999 See page 4, line 15 - line 25, page 7, line 15- page 8, line 24, page 9, line 20 - page 10, line 20	1, 3, 4, 5, 6, 11
Y	US, A, 6 147 009 (IBM) 14 November 2000 See the whole document	1
Y	US A, 6 077 574 (NEC CORP.) 20 June 2000 See the whole document	1

☐ Further documents are listed in the continuation of Box C.

☐ See patent family annex.

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Date of the actual completion of the international search

29 AUGUST 2002 (29.08.2002)

Date of mailing of the international search report

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